Magnetic order in the two-dimensional compass-Heisenberg model

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A Green-function theory for the dynamic spin susceptibility in the square-lattice spin-1/2 antiferromagnetic compass-Heisenberg model employing a generalized mean-field approximation is presented. The theory describes magnetic long-range order (LRO) and short-range order (SRO) at arbitrary temperatures. The magnetization, Néel temperature T_N , specific heat, and uniform static spin susceptibility χ are calculated self-consistently. As the main result, we obtain LRO at finite temperatures in two dimensions, where the dependence of T_N on the compass-model interaction is studied. We find that T_N is close to the experimental value for Ba_2IrO_4 . The effects of SRO are discussed in relation to the temperature dependence of χ .

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I. INTRODUCTION

The study of the quantum compass model (CM) for strongly correlated transition-metal compounds with orbital degrees of freedom and strong spin-orbit coupling is an active field of research (for a review, see Ref. [1]). In particular, quantum and thermodynamic phase transitions in the two-dimensional (2D) CM were studied in Refs. [2, 3], where the directional-ordering transition of the 2D Ising universality class was found for the symmetric CM. Depending on the method of numerical computation, the temperature of the phase transition for the quantum CM was found in the range $T_c = 0.055 - 0.058$ (in the units of the CM exchange interaction) [3]. In Refs. [4, 5] the compass-Heisenberg (CH) model was introduced extending the CM by the 2D antiferromagnetic (AF) Heisenberg model, and the ground state and excited states of the CH model were analyzed. For 5d transition-metal compounds with a strong spin-orbit coupling, such as Sr_2IrO_4 and Sr_2IrO_4 with the Néel temperatures $T_N = 230K$ and $T_N = 240K$, respectively, (see, e.g., Ref. [6]), an effective AF Heisenberg model for pseudospins S = 1/2 with the CM anisotropy was derived in Ref. [7]. Recently we have calculated the spin-wave excitation spectrum and T_N for a layered CH model by means of the random phase approximation (RPA) [8] (see also Refs. [9]). An important issue is the description of magnetic long-range order (LRO) and short-range order (SRO) and of the thermodynamics at arbitrary temperatures by a theory going beyond RPA (SRO) and of the thermodynamics at arbitrary temperatures by a theory going beyond RPA.

In this paper we employ a generalized mean-field approximation (GMFA) to the 2D CH model that is based on the equation-of-motion method for Green functions. In the framework of a more general theory for the dynamic spin susceptibility including the self energy [10, 11], the neglect of the self-energy corresponds to the GMFA. This approximation, for spin-rotation invariant (SRI) systems also named SRI Green-function method (RGM), has been successfully applied to several quantum spin systems (see, e.g., Refs. [12–26]).

We start from the spin-1/2 CH model on the square lattice,

$$H = \frac{1}{2} \sum_{i,j,\nu} J_{ij}^{\nu} S_i^{\nu} S_j^{\nu}, \tag{1}$$

where $\nu = x, y, z$. The nearest-neighbor (NN) exchange interaction parameters are $J_{ij}^x = J_{ij} + \Gamma_{ij}^x$, $J_{ij}^y =$ $J_{ij} + \Gamma^{y}_{ij}, J^{z}_{ij} = J_{ij}, J_{ij} = J(\delta_{\mathbf{r}_{j},\mathbf{r}_{i}\pm\mathbf{a}_{x}} + \delta_{\mathbf{r}_{j},\mathbf{r}_{i}\pm\mathbf{a}_{y}}),$ $\Gamma^{x}_{ij} = \Gamma_{x}\delta_{\mathbf{r}_{j},\mathbf{r}_{i}\pm\mathbf{a}_{x}}, \text{ and } \Gamma^{y}_{ij} = \Gamma_{y}\delta_{\mathbf{r}_{j},\mathbf{r}_{i}\pm\mathbf{a}_{y}}.$ We assume J > 0 and $\Gamma_{x} \geq \Gamma_{y} > 0$. The symmetric formulation of the model (1) allows us to get expressions for quantities with indexes $\nu = y$, z from those indicated by the index $\nu = x$ by cyclic permutation.

THEORY OF SPIN SUSCEPTIBILITY

To evaluate the thermodynamic quantities in the CH model, we calculate the dynamic spin susceptibility $\chi^{\nu}_{\mathbf{q}}(\omega) = -\langle\langle S^{\nu}_{\mathbf{q}}|S^{\nu}_{\mathbf{q}}\rangle\rangle_{\omega} \ (\langle\langle...|...\rangle\rangle_{\omega} \ \text{denotes the retarded}$ two-time commutator Green function [27]). Using the equations of motion up to the second step, we obtain $\omega^2 \langle \langle S_{\mathbf{q}}^{\nu} | S_{\mathbf{q}}^{\nu} \rangle \rangle_{\omega} = m_{\mathbf{q}}^{\nu} + \langle \langle -\ddot{S}_{\mathbf{q}}^{\nu} | S_{\mathbf{q}}^{\nu} \rangle \rangle_{\omega}$, where $m_{\mathbf{q}}^{\nu} = \langle [i\dot{S}_{\mathbf{q}}^{\nu} | S_{\mathbf{q}}^{\nu}] \rangle$, $i\dot{S}_{\mathbf{q}}^{\nu} = [S_{\mathbf{q}}^{\nu}, H]$, and $-\ddot{S}_{\mathbf{q}}^{\nu} = [[S_{\mathbf{q}}^{\nu}, H], H]$. For the model (1) the moment $m_{\mathbf{q}}^{x}$ is given by the exact ex-

$$m_{\mathbf{q}}^{x} = \sum_{i} [\cos(\mathbf{q}\mathbf{R}_{i})(J_{0i}^{y}C_{0i}^{z} + J_{0i}^{z}C_{0i}^{y}) - J_{0i}^{y}C_{0i}^{y} - J_{0i}^{z}C_{0i}^{z}].$$
(2)

Here, $C_{ij}^{\nu} = C_{\mathbf{R}_{j}-\mathbf{R}_{i}}^{\nu} = \langle S_{0}^{\nu} S_{\mathbf{R}_{j}-\mathbf{R}_{i}}^{\nu} \rangle$ with $\mathbf{R} = m\mathbf{e}_{\mathbf{x}} + n\mathbf{e}_{\mathbf{y}}$ denote the spin correlation functions.

The second derivatives $-\ddot{S}^{\nu}_{\mathbf{q}}$ are approximated in the spirit of the scheme employed in Refs. [12–26]. That means, taking the site representation, in $-\ddot{S}_i^x$ we decouple the products of three spin operators on different lattice sites along NN sequences $\langle i, j, k \rangle$ as

$$S_i^x S_i^y S_k^y = \alpha_1^x \langle S_i^y S_k^y \rangle S_i^x, \tag{3}$$

$$S_i^x S_i^y S_k^y = \alpha_2^x \langle S_i^y S_k^y \rangle S_i^x, \tag{4}$$

where the vertex renormalization parameters α_1^x and α_2^x are attached to NN and further-distant correlation functions, respectively. After some algebra, we obtain $-\ddot{S}_{\mathbf{q}}^{\nu} = (\omega_{\mathbf{q}}^{\nu})^2 S_{\mathbf{q}}^{\nu}$ and

$$\chi_{\mathbf{q}}^{\nu}(\omega) = -\langle\langle S_{\mathbf{q}}^{\nu} | S_{\mathbf{q}}^{\nu} \rangle\rangle_{\omega} = \frac{m_{\mathbf{q}}^{\nu}}{(\omega_{\mathbf{q}}^{\nu})^{2} - \omega^{2}}, \tag{5}$$

with the squared spin-excitation energy

$$(\omega_{\mathbf{q}}^x)^2 = \sum_{i,j} [\delta_{ij} a_i^x(\mathbf{q}) + (1 - \delta_{ij}) b_{ij}^x(\mathbf{q})], \tag{6}$$

where

$$a_i^x(\mathbf{q}) = \frac{1}{4} [(J_{0i}^y)^2 + (J_{0i}^z)^2 - 2J_{0i}^y J_{0i}^z \cos(\mathbf{q}\mathbf{R}_i)], \qquad (7)$$

$$b_{ij}^{x}(\mathbf{q}) = \alpha_{2}^{x} J_{0i}^{y} J_{0j}^{y} C_{ij}^{y} + \alpha_{2}^{x} J_{0i}^{z} J_{0j}^{z} C_{ij}^{z}$$

$$+ \alpha_{1}^{x} \cos(\mathbf{q} \mathbf{R}_{ij}) (J_{0i}^{y} J_{0j}^{x} C_{0i}^{z} + J_{0i}^{z} J_{0j}^{x} C_{0i}^{y})$$

$$- \cos(\mathbf{q} \mathbf{R}_{i}) (\alpha_{1}^{x} J_{0i}^{x} J_{0j}^{y} C_{0j}^{y} + \alpha_{1}^{x} J_{0i}^{x} J_{0j}^{z} C_{0j}^{z}$$

$$+ \alpha_{2}^{x} J_{0i}^{y} J_{0j}^{z} C_{ij}^{z} + \alpha_{2}^{x} J_{0i}^{z} J_{0j}^{y} C_{ij}^{y}).$$

$$(8)$$

The appearance of AF LRO at $T \leq T_N$ is reflected in our theory by the divergence of $\chi^{\nu}_{\mathbf{Q}} \equiv \chi^{\nu}_{\mathbf{Q}}(\omega=0)$ corresponding to the closure of the spectrum gap at the AF ordering vector $\mathbf{Q} = (\pi, \pi)$, $\omega^{\nu}_{\mathbf{Q}}(T \leq T_N) = 0$. In the LRO phase the correlation functions $C^{\nu}_{\mathbf{R}} \equiv C^{\nu}_{m,n}$ are written as [13, 15, 16, 22, 25, 26]

$$C_{\mathbf{R}}^{\nu} = \frac{1}{N} \sum_{\mathbf{q} \neq \mathbf{Q}} C_{\mathbf{q}}^{\nu} e^{i\mathbf{q}\mathbf{R}} + C^{\nu} e^{i\mathbf{Q}\mathbf{R}}, \tag{9}$$

with $C_{\mathbf{q}}^{\nu}$ calculated from the Green function (5) by the spectral theorem,

$$C_{\mathbf{q}}^{\nu} = \langle S_{\mathbf{q}}^{\nu} S_{\mathbf{q}}^{\nu} \rangle = \frac{m_{\mathbf{q}}^{\nu}}{2\omega_{\mathbf{q}}^{\nu}} [1 + 2n(\omega_{\mathbf{q}}^{\nu})], \tag{10}$$

where $n(\omega) = (e^{\omega/T} - 1)^{-1}$. The condensation part C^{ν} arising from $\omega_{\mathbf{Q}}^{\nu} = 0$ determines the staggered magnetization m^{ν} that is defined by

$$(m^{\nu})^2 = \frac{1}{N} \sum_{\mathbf{R}} C_{\mathbf{R}}^{\nu} e^{-i\mathbf{Q}\mathbf{R}} = C^{\nu}.$$
 (11)

In the paramagnetic phase, we have $\omega_{\mathbf{Q}}^{\nu} > 0$ and $C^{\nu} = 0$. The NN correlation functions are related to the internal energy u per site, $u = (1/2N) \sum_{i,j,\nu} J_{ij}^{\nu} C_{ij}^{\nu}$, from which the specific heat $C_V = du/dT$ may be obtained.

To calculate the thermodynamic properties, the correlation functions $C_{\mathbf{R}}^{\nu}$, the vertex parameters $\alpha_{1,2}^{\nu}$ appearing in the spectrum $\omega_{\mathbf{q}}^{\nu}$, and the condensation term C^{ν} in the LRO phase have to be determined. Besides Eqs. (9) and (10) for calculating the correlators, we have the sum rules $C_{\mathbf{R}=0}^{\nu}=1/4$ and the LRO conditions $\omega_{\mathbf{Q}}^{\nu}=0$; that is, we have more parameters than equations. To obtain a closed system of self-consistency equations, we

proceed as follows. As in our recent study of the model (1) by means of the RPA and linear spin-wave theory (LSWT) [8], we consider an anisotropic CM interaction, $\Gamma_x > \Gamma_y > 0$, so that the LRO phase is an easy-axis AF with the magnetization along the x axis. Accordingly, we put $C^y = C^z = 0$, where we can also consider the limiting case $\Gamma_x = \Gamma_y$. Then, at T = 0, for determining the seven quantities $\alpha_{1,2}^{\nu}$ and C^{x} , besides the three sum rules and the three LRO conditions, we need an additional condition. To this end, we adjust the ground-state magnetization $m^{x}(0)$ to the expression obtained in LSWT, $m^{x}(0) = m_{LSWT}^{x}(0) = (1/2) - (1/N) \sum_{\mathbf{q}} \langle S_{\mathbf{q}}^{-} S_{\mathbf{q}}^{+} \rangle$, where the correlation function is given by Eq. (17) of Ref. [8] with the sublattice magnetization σ substituted by the spin S = 1/2. In the LRO phase, $0 < T \le T_N$, we have found that the ansatz $\bar{r}_{\alpha}(T) = \bar{r}_{\alpha}(0)$, where $\bar{r}_{\alpha} = (r_{\alpha}^x r_{\alpha}^y r_{\alpha}^z)^{1/3} \text{ with } r_{\alpha}^{\nu}(T) = (\alpha_2^{\nu}(T) - 1)/(\alpha_1^{\nu}(T) - 1),$ is a reasonable approximation, as will be demonstrated in the Appendix for the Heisenberg limit. At $T > T_N$, for calculating $\alpha_{1,2}^{\nu}$ we use the sum rules and the condition $r^{\nu}_{\alpha}(T) = r^{\nu}_{\alpha}(T_N).$

Because, as an input, we take the ground-state magnetization in LSWT that describes the LRO quite well for small enough CM interaction $\Gamma_{x,y}$ as compared to the exchange interaction J, we present results in the region $\Gamma_{x,y}/J \ll 1$.

III. RESULTS

As described in Sec. II, the thermodynamic quantities are calculated from the numerical solution of a coupled system of nonlinear algebraic self-consistency equations.

First we consider our results on the magnetic LRO. In the Heisenberg limit we have no LRO at finite temperature, in accord with the Mermin-Wagner theorem. In Fig. 1 the Néel temperature T_N as function of Γ_y for two ratios of Γ_x/Γ_y is plotted. Our results for T_N remarkably deviate from those found in RPA [8]. For $\Gamma_x/\Gamma_y = 1.5$, T_N exhibits qualitatively the same dependence on Γ_y , but is appreciably reduced as compared to RPA. For the symmetric CM interaction $\Gamma_x = \Gamma_y = \Gamma$, our theory yields a finite Néel temperature that increases with Γ , whereas in RPA, $T_N = 0$ was found. We ascribe these differences to a better description of strong spin fluctuations by our Green-function theory going one step beyond RPA. Note that in the RPA [8] the fluctuations of the x component of spin are not taken into account. Generally speaking, from our results we conclude that the CM interaction added to the Heisenberg model favors magnetic LRO at finite T in two dimensions.

Comparing our findings to experiment, we consider the compound Ba₂IrO₄, where quantum chemistry calculations [28] yield the parameters J=65meV, $\Gamma=3.4$ meV = 0.0523J, and a very small interplane coupling $J_z\approx 5\times 10^{-5}J$ that is not taken into account here. In Fig. 2 the magnetization $m=m^x$ as function of T is plotted. At T=0, m is enhanced by Γ as compared to the value in

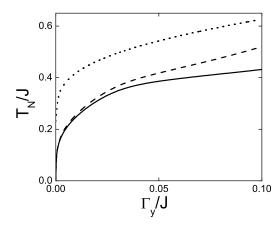


FIG. 1: Néel temperature T_N for $\Gamma_x = \Gamma_y$ (solid) and $\Gamma_x = 1.5\Gamma_y$ (dashed) compared with the RPA result (dotted).

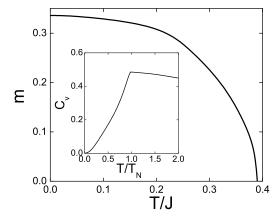


FIG. 2: Magnetization m and specific heat C_V for $\Gamma_x = \Gamma_y = 0.0523J$.

the Heisenberg limit $(m_{LSWT}(0) = 0.303)$. The second-order phase transition occurs at $T_N = 0.392J = 295 \mathrm{K}$ which agrees rather well with the experimental value $T_N^{exp} = 240 \mathrm{K}$. At T_N the specific heat (inset to Fig. 2) reveals a cusplike singularity. Such a structure at the transition temperature was also found in layered magnets treated by RGM in Ref. [22]. From the analysis given there we suggest that also here the height of the cusp may be underestimated. It is desirable to compare our result for C_V to experimental data which, however, are not yet available.

Finally, we discuss the uniform static spin susceptibility $\chi(T) \equiv \chi_{{\bf q}=0}(\omega=0)$ depicted in Fig. 3 for the symmetric CM interaction, which reflects the behavior of magnetic SRO. Considering the Heisenberg limit, the increase of χ with temperature is caused by the decrease of AF SRO, i.e., of the spin stiffness against the orientation along a homogeneous magnetic field. Since the SRO is less pronounced at higher T, χ reveals a maximum near the exchange energy and a crossover to the high-temperature Curie-Weiss behavior. In the paramagnetic phase for $\Gamma>0$, the susceptibilities χ^{ν} are low-

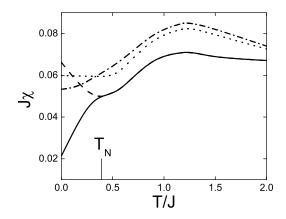


FIG. 3: Uniform static susceptibility χ for $\Gamma_x = \Gamma_y = 0.0523J$, χ^x (solid), χ^y (dashed), χ^z (dotted), and for the Heisenberg limit (dot-dashed).

ered as compared with the Heisenberg limit due to the Γ -induced enhancement of AF SRO. Moreover, the susceptibility $\chi^x = \chi^y$ is reduced as compared with χ^z , because the SRO of the x- and y-spin components is more pronounced due to the AF CM interaction along the x- and y-axes. In the LRO phase, the susceptibility χ^x strongly decreases with decreasing T, because the increasing easy-x-axis LRO also enhances the SRO. Considering the susceptibility χ^y , with decreasing T below T_N the SRO of the y-spin components is reduced due to the increasing alignment of the spins in x-direction, so that χ^y increases with decreasing T. Thus, χ^y exhibits a minimum at T_N . The susceptibility χ^z in the LRO phase turns out to be nearly temperature independent.

IV. SUMMARY

In this paper we have calculated thermodynamic properties of the 2D antiferromagnetic CH model within a generalized mean-field approach for arbitrary temperatures. Our main focus was the detailed investigation of magnetic LRO. We have found that the Néel temperature T_N is finite even in the case of a symmetric CM interaction. We have explained the temperature dependence of the uniform static susceptibility in terms of magnetic SRO. Our investigation forms the basis for forthcoming extended studies of the CH model. Especially, the generalization of the theory holding for arbitrary CH model parameters and allowing for the description of magnetic and directional-ordering transitions is of particular interest.

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TABLE I: Heisenberg limit at T = 0.

	$\nu = x$	$\nu = y, z$	average	SRI
C_{10}	-0.1542	-0.0989	-0.1173	-0.1173
C_{20}	0.118	0.0412	0.0668	0.0667
C_{11}	0.125	0.0522	0.0765	0.0763
$J\chi$	0.0327	0.0636	0.0533	0.05292
Ω/J	3.477	2.821	3.04	2.978

Appendix A: Heisenberg limit

In the limit $\Gamma_{x,y} = 0$, the LRO in the ground state of the Heisenberg AF may be described within the RGM by the SRI forms of Eqs. (9) and (11), i.e., by $C_{\mathbf{R}}^x = C_{\mathbf{R}}^y = C_{\mathbf{R}}^z$ and $C^x = C^y = C^z$ as it was done, e.g., in Refs. [13, 15, 22, 26]. According to the consideration of the CH model with the easy-axis magnetization m^x , let us outline an alternative possibility to describe the ground-state LRO in the AF Heisenberg model. Here, we may also break the rotational symmetry by putting $C^y = C^z = 0$, which is analogous to the introduction of a symmetry-breaking sublattice magnetization (see, e.g.,

our RPA approach [8]). Performing the calculations at T=0 as described in Sec. II, we obtain the correlation functions C_{mn}^{ν} and the uniform static spin susceptibility χ^{ν} listed in Table I. The spin-excitation spectrum at T=0 is found to be

$$\omega_{\mathbf{q}}^{\nu} = \Omega^{\nu} \sqrt{1 - \gamma_{\mathbf{q}}^2},\tag{A1}$$

where $\gamma_{\bf q}=\frac{1}{2}(\cos q_x+\cos q_y)$ and $(\Omega^x)^2=-16J^2\alpha_1^x(C_{10}^y+C_{10}^z)$. The amplitudes Ω^ν are given in Table I. Note that Eq. (A1) leads to the SRI result (Ref. [15]), if we put $\alpha_1^x=\alpha_1$ and $C_{10}^y+C_{10}^z=2C_{10}^z$, and has the same shape as in LSWT and RPA.

To relate the non-SRI description of LRO to the SRI formulation, we calculate arithmetic averages, e.g., $C_{mn} = \frac{1}{3} \sum_{\nu} C_{mn}^{\nu}$. As can be seen from Table I, we obtain a very good agreement of the averaged ground-state properties with those resulting from the SRI theory. Moreover, the geometrical average $\bar{r}_{\alpha}(0)$ used in our ansatz for the LRO phase (see Sec. II) given in the Heisenberg limit by $\bar{r}_{\alpha}(0) = 1.1913$ nearly agrees with the ratio $r_{\alpha}(0) = 1.2109$ obtained in the SRI approach (see also Ref. [13]). This gives some justification for formulating our ansatz in terms of the geometrical average.

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